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ABSTRACT

Small micrometer-sized roughness on optical surfaces, caused by laser damage and/or redeposition of laser ablated material, can cause local electric field intensification which may lead to damage initiation both on the optics and/or downstream. We examined the smoothing of etched periodic surface structures on SiO_2 substrate with 10.6μm CO_2 laser using atomic force microscopy. The characteristic surface tension driven mass flow of the glass under different laser parameters were simulated using computational fluid dynamics and correlated with experimental results. We found that during CO_2 laser polishing the estimate viscosity of the silica glass appears to be higher than typical literature values measured at a temperature similar to the laser heating conditions. This discrepancy can be explained by the observation that at high temperature, a significant portion of the hydroxyl content in the layer of heated silica glass can diffuse out resulting in a much stiffer glass.
Keywords: Laser polishing, capillarity, fused silica, viscosity, hydroxyl content, damage mitigation, CO_2 laser

1. INTRODUCTION

CO_2 laser heating of silica glass has been used to control and mitigate laser induced damage for high average power laser applications such as inertial confinement fusion (ICF) [1, 2]. In particular, we are interested in minimizing damage initiation on optics surface and/or downstream caused by small-scale surface roughness resulting from laser machining or optical damage [3-5]. Figure 1a shows a typical AFM image of debris on the optics surface after a laser machining protocol. The surface roughness causes perturbation to the laser field propagation and may lead to field intensification as shown by the electromagnetic field simulation in figure 1(b).

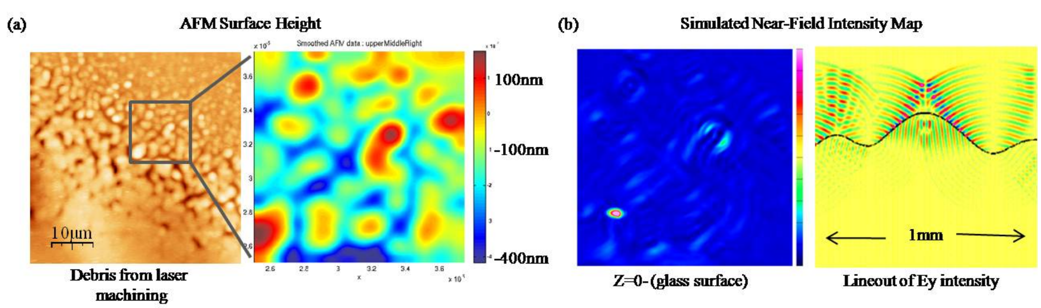

Figure 1. AFM scan of a fused silica sample after laser machining and the corresponding field intensity map from the laser beam propagation simulation.

The CO_2 laser locally heats up the glass and allows the rough surface to relax. The processes that govern the relaxation of a heated surface S in an amorphous material such as glass are viscous flow, evaporation-condensation, bulk diffusion, and surface diffusion [3-8]. Mullin has outlined the relative importance of the four transport processes,

$$\frac{\partial S}{\partial t} = -(F\omega + A\omega^2 + D\omega^3 + B\omega^4)S \tag{1}$$

where F, A, D, and B are the decay constants for the four processes. Here, we focus on features ~1um in size. For the relaxation of these surface features, surface diffusion and viscous flow should be the dominant transport processes. However, it has been demonstrated by isothermal heating of grating structures on glasses that the effect of surface diffusion is negligible [9]. One possible explanation the author attributed to the observation is the existence of defects which reduces surface diffusion [10].

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To better predict the effect of CO_2 laser polishing/mitigation of silica surface roughness, we simulate the process of viscous glass flow using computational fluid dynamics (CFD) software and compare it to experimental results obtained from laser polishing of one dimensional gratings etched on fused silica glass.

2. SIMULATION OF SURFACE RELAXATION

We used commercially available computational fluid dynamics (CFD) software, Flow3D, to simulate the surface tension driven mass flow of heated glass under the CO_2 laser smoothing conditions achieved in our experiments. For the system we are considering, the inertial and gravitational effects are negligible. The smoothing flow rate is determined by the ratio of the capillary force and viscous force. Therefore, to simplify the simulation, we define a non-dimensional variable τ as the ratio of the Reynolds number and the Weber number

$$\tau = \frac{N_{Re}}{N_{We}} = \frac{\sigma}{\mu(T)V_0} = \frac{\sigma t}{\mu(T)w} \qquad (2)$$

where σ is the surface tension, μ is the temperature dependent viscosity, t is the laser exposure time, and w is the width of the grating groove. The surface tension σ is taken as a constant of 300 dyne/cm over the range of temperature in our experimental system. The parameter τ characterizes the "normalized" time that the relaxation of a surface feature w with surface tension σ and viscosity μ (T). By using τ as a variable in the simulation, we collapse the feature height vs. time plots at different temperature onto a single curve, greatly reducing modeling time. We also assumed free slip boundary conditions with 0 flux. The contact angles at the peak and valley of the grating were kept at 90˚.

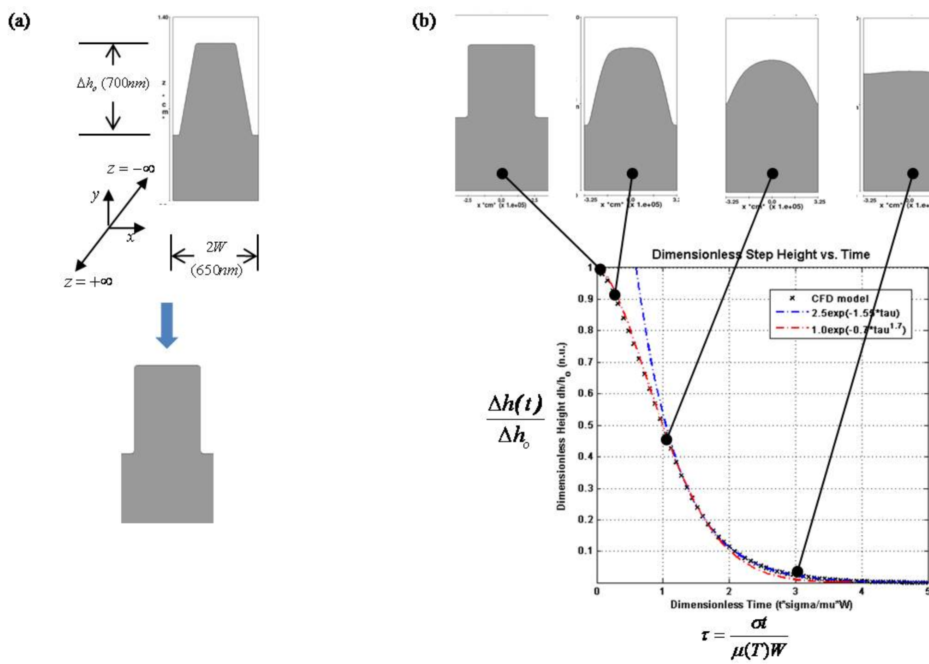

Figure 2. Computation fluid mechanics simulation of the relaxation of a grating structure. (a) A square shape is used to approximate the trapezoidal structure; (b) functional fits of the simulation.

The normalized height $\Delta h(t)/\Delta h_0$ of the grating is plotted as a function of τ in Figure 2. Although the grating we are simulating does not have a perfect square shape, it decays very similarly to a perfect square feature. We therefore took the analytical expressions that describe the relaxation of a square structure and compared it with our experimental results. For τ<1.5, the edge effect of the square feature is significant. It takes a modified exponential to describe the smoothing behavior. Once the surface feature assumes a pure sinusoidal shape, it decays exponentially as a function of τ.

$$\frac{\Delta h(t)}{\Delta h_0} = e^{-0.7\tau^{1.7}} \qquad \text{for } \tau<1.5 \qquad (3)$$

$$\frac{\Delta h(t)}{\Delta h_0} = 2.5e^{-1.55\tau} \qquad \text{for } \tau\geq1.5 \qquad (4)$$

3. SAMPLE PREPARATION AND EXPERIMENTS

We used two types of fused silica glass, Corning 7980, Heraeus Suprasil 312, and BK7 in the study. The two silica glass types differ in their residual water content which we verified using micro-Raman measurement [11]. One-dimensional gratings with a wavelength of ~650nm and a height of ~700nm were patterned on 2 inch round silica samples using laser interference lithography and ion-beam etching [12]. The surface morphology of the etched samples was characterized using a Digital Instrument Dimension 3100 AFM (Figure 3). We used high aspect ratio silicon tips (Veeco OTESPAW) to ensure the accuracy of our AFM results.

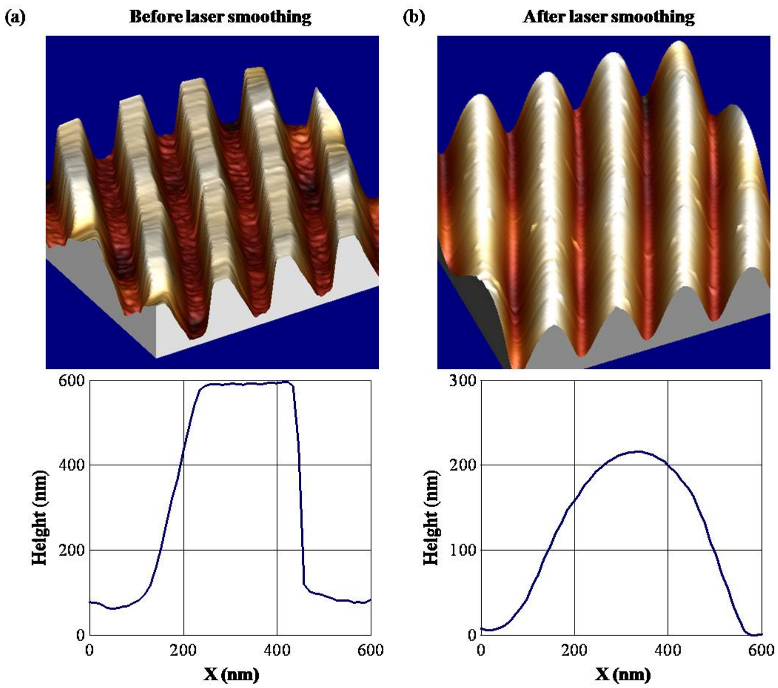

Figure 3. Typical AFM images of grating (a) before and (b) after CO_2 laser polishing.

The silica and BK7 samples were treated with single CW 10.6μm CO_2 laser beam from a Synrad firestar V20, with a maximum output power of 20 watts and power stability of ±5%. The laser beam profile was nearly a perfect Gaussian a $1/e^2$ diameter of ~1mm, as characterized by a beam profiler (Pyrocam III, Spiricon). During the laser polishing process, we monitored the sample surface temperature in-situ by using a calibrated thermographic camera [13]. We varied the laser power from 3.8 to 4.7W for the fused silica samples and 1 to 1.4W for the BK7 sample. The laser exposure time ranged from 100ms to 100s. The surface morphology of the samples was measured again using AFM after laser polishing.

4. RESULTS

The spatial profile of the laser power translates to a variation in surface temperature resulting in an effective viscosity profile (Figure 4). As a result, by measuring the height change of the grating at different locations within one laser beam, we were able to assess laser smoothing effects for a range of temperature, hence viscosity. Since we are studying the behavior of glass flow in a relatively small temperature range, the temperature dependant viscosity can be described by an Arrhenius expression [14]

$$\mu = \mu_0 e^{\left(\frac{E_a}{RT}\right)} \quad (5)$$

where E_a is the activation energy of viscous glass flow.

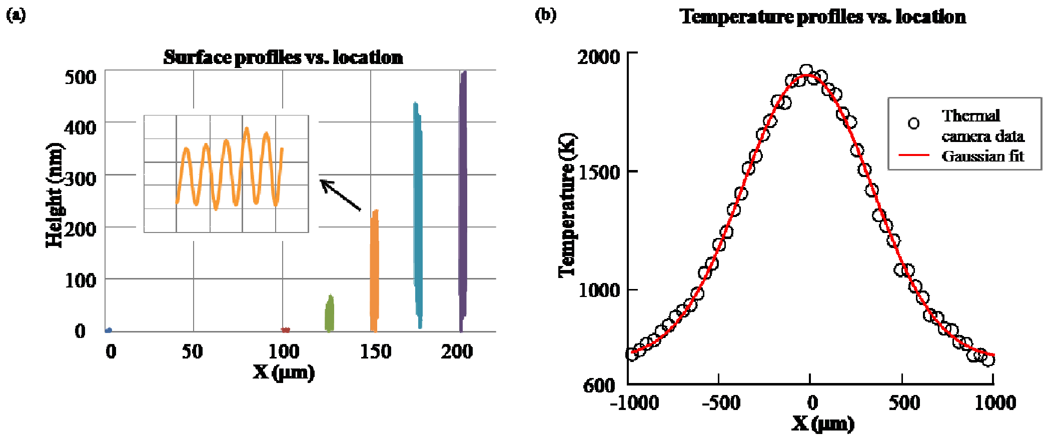

Figure 4. Spatial map of the region where surface profiles of the grating were measured using AFM and the corresponding temperature at that location. (a) Microscopy image of region treated with the Gaussian CO_2 laser beam; (b) the grating height at different locations relative to laser beam center, and (c) the in-situ temperature measurement.

Based on the surface temperature measurement from the thermographic camera, we could calculate the viscosity value for a glass using equation (5) given that we know the values of μ_0 and E_a for that type of glass. Corning 7980 fused silica is a type III glass containing ~1000ppm OH with a measured value of μ_0=8.81×10^{-5} Poise and E_a=434 kJ/mole [15, 16]. When these numbers are used in equation (5) for the 3.8W, 100s exposure condition and compared it with our CFD simulation, we found big discrepancy between the two (Figure 4). Using values of μ_0 and E_a for type II glass (~150ppm OH content) in the calculation brought the two curves closer but still not satisfactory.

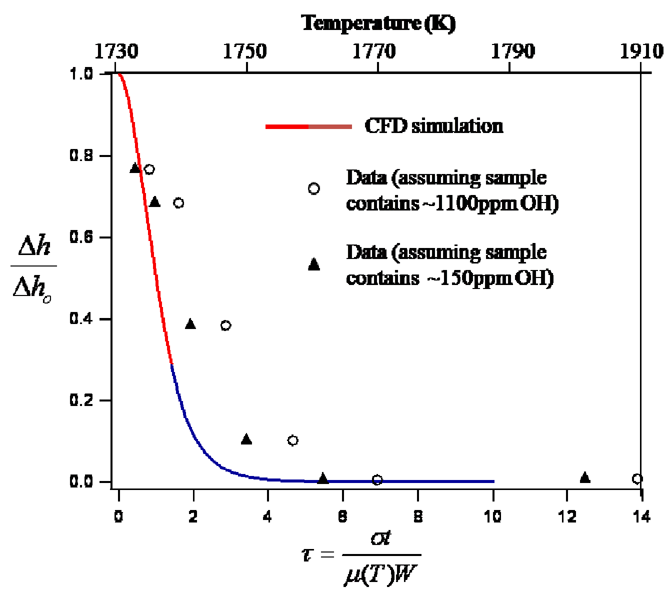

Figure 5: Experiential and simulation results of normalized grating height vs. τ. The values of τ was calculated using Arrhenius relations for viscosity reported in the literature for silica glasses with ~1100 OH content (circles) and ~150ppm OH content (triangles).

In the experiment, we control the laser exposure time and can directly measure the grating structure height and the sample surface temperature. The only unknown parameter is the temperature dependent viscosity. By fitting $\Delta h(t)/\Delta h_0$ as a function of T, we obtained the Arrhenius expression of the viscosity for Corning 7980 glass under CO_2 laser polishing condition (Figure 5a). A good agreement between the experiment and simulation is achieved with several different laser parameters when we used this new Arrhenius expression for viscosity (Figure 5b). The higher activation energy E_a suggests that the hydroxyl content may have been diffused out during laser heating. This observation has been reported also by Wang et al. [9].

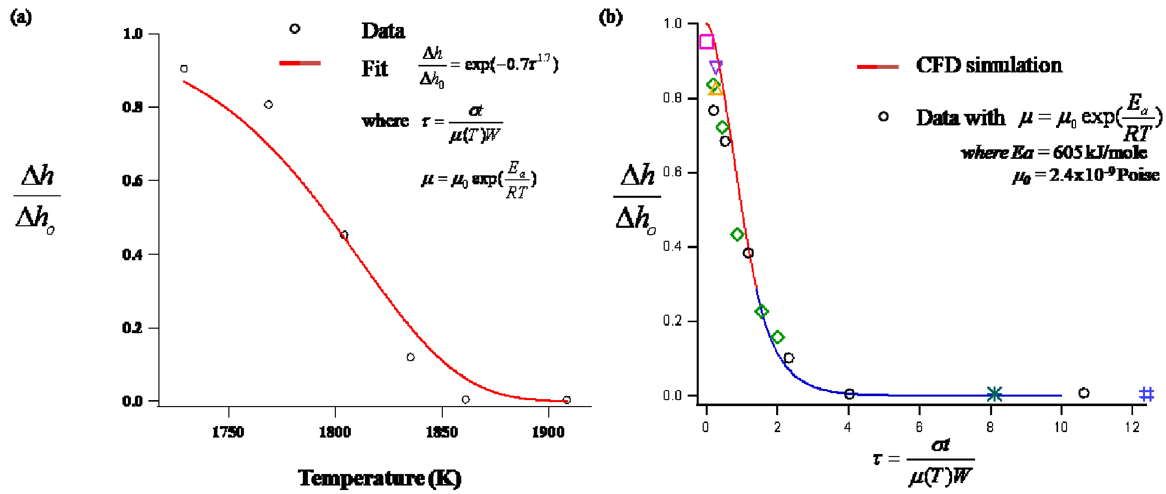

Figure 6. Arrhenius viscosity relation for type II silica glass and its smoothing under different laser parameters. (a) time dependent viscosity of the silica glass under the CO_2 laser polishing condition; (b) grating height evolution as a function of τ with varying laser power and time.

We analyzed the effect of laser polishing of type II fused silica glass (Heraeus Suprasil 312) using the same method. The result also showed a depletion of OH during laser heating. Table 1 summarizes the values of the activation energy for the two types of silica glass and BK7 found in the literature [15-17], and the values measured under our experimental conditions.

Table I. Activation energy of viscous flow for different fused silica glass.

Sample	Reported activation energy (kJ/mole)	Measured activation energy (kJ/mole)
Corning 7980	434	605
BK7	96	162
Suprasil 312	515	578

5. DISCUSSION

Although we simplified the problem of surface relaxation by considering only the heat activated surface tension driven mass flow, the model adequately describes the CO_2 laser polishing of micrometer sized roughness in fused silica glass. The computational fluid dynamics simulation based on this model showed good agreement with experimental data. It is worth noting that the non-dimensional parameter τ gives some insight on the relationship between time and viscosity (temperature) during laser polishing. When designing protocols for polishing a feature of size *w*, it is possible to achieve smoothing by either picking the right time *t* or temperature to reach a desired viscosity.

Our study verifies the finding from other groups that during heating of fused silica glass, the OH content depletion occurs inside the surface layer. We performed Raman spectroscopy to monitor the OH variation on the sample surface and in bulk. The Si-OH stretch at 970cm^{-1} gives reasonable estimate of the amount of OH [9]. The estimated bulk OH contents are ~1000ppm for the Corning 7980 and ~200ppm for Heraeus Suprasil 312, close to the values specified by the vendors. One note worth mentioning is that the small τ data (for lower laser power and/or shorter exposure time) in Figure 5b did not agree with the simulation as well as the larger τ data. We suspect that the OH depletion may not be uniform with respect to laser parameter. Further experiment is planned to better understand the phenomena.

Viscosity limits the relaxation processes near glass transition temperature. Therefore, knowing the viscosity of a glass is important to predicting processing results. Direct measurement of the viscosity of glass at high temperature is often difficult, especially under extreme conditions such as CO_2 laser mitigation. Study of the relaxation of grating structures may provide an attractive method to indirectly fine-tune the temperature dependant viscosity at high temperature.

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